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14. ABSTRACT

The importance of auto- and forced ignition in the operation of practical combustion devices such as the Diesel engine suggests that better understanding and enhanced performance of these devices rely on knowledge of the separate and coupled roles of the complex chemical kinetics and unsteady and turbulent aerodynamics that are present in such systems. This investigation adopted the counterflow configuration, with its well defined flow time, and employed laser-based experimentation, computational simulation with detailed chemistry and transport, turbulent modeling, and analysis with reduced mechanisms in an attempt to achieve a comprehensive understanding of the various controlling processes and parameters. The chemical kinetic portion involved the experimental determination of the ignition temperature as a function of the fuel concentration, system pressure, and flow strain rate for selected hydrocarbons up to C_8, and the subsequent modeling and development of detailed, simplified, and reduced kinetic mechanisms governing ignition of these fuels. The aerodynamic portion of the program involved experimental measurement and numerical modeling of the flow field present immediately prior to ignition in a turbulent counterflow, as well as the determination of ignition temperatures as a function of turbulent intensity and the other parameters investigated in laminar flows.

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STATEMENT OF PROBLEM

The importance of auto- and forced ignition in the operation of practical combustion devices such as the Diesel engine suggests that better understanding and enhanced performance of these devices rely on knowledge of the separate and coupled roles of the complex chemical kinetics and unsteady and turbulent aerodynamics that are present in such systems. This investigation adopted the counterflow configuration, with its well defined flow time, and employed laser-based experimentation, computational simulation with detailed chemistry and transport, turbulent modeling, and analysis with reduced mechanisms in an attempt to achieve a comprehensive understanding of the various controlling processes and parameters. The chemical kinetic portion involved the experimental determination of the ignition temperature as a function of the fuel concentration, system pressure, and flow strain rate for selected hydrocarbons up to C_8, and the subsequent modeling and development of detailed, simplified, and reduced kinetic mechanisms governing ignition of these fuels. The aerodynamic portion of the program involved experimental measurement and numerical modeling of the flow field present immediately prior to ignition in a turbulent counterflow, as well as the determination of ignition temperatures as a function of turbulent intensity and the other parameters investigated in laminar flows.

RESEARCH SUMMARY

1. Ignition of n-heptane and iso-octane
N-heptane and iso-octane are used as reference fuels for engine knock because they have similar vaporization pressures while their fuel structures are representative of two important constituents of practical fuels, linear and branched alkanes. Laminar nonpremixed ignition experiments were conducted for n-heptane and iso-octane and the air temperature required to ignite the fuel mixture is indicated as a functions of strain rate in Fig. 1 and pressure in Fig. 2. Since ignition in this configuration provides an additional test for the comprehensiveness of chemical models due to the highly non-uniform conditions experienced by the reactants as they are swept through the flow field, existing empirical models were compared with the experiments.

Ignition temperatures increased for both fuels as the strain rate increased due to reduced residence time. Increased pressure caused the ignition temperatures to decrease for both fuels because the limiting reactions are facilitated at higher pressures whereas the pressure-weighted diffusion rates are not affected much. N-heptane ignited at lower temperatures than iso-octane, consistent with the fact that its alkyl radical is much less reactive, resulting in a net consumption of hydrogen radicals from hydrogen abstraction. All of these results are consistent with the ignition of smaller hydrocarbon fuels.

Using the semi-empirical kinetic models, the calculated ignition temperatures were about 100K higher than the experiments, similar to the discrepancy found when modeling methane and ethane ignition. Diffusive sensitivity analysis indicated that fuel diffusivity is very important, but even a 50% increase in diffusivity does not bring the experiments and calculations into agreement. All reactions have lower sensitivities than fuel diffusion, and the highest reaction sensitivity coefficients are associated with C_3

species and smaller. Both heat release and chain branching are necessary to achieve ignition. These computational results are also similar to results for ethane.

The models do not include elements of low temperature chemistry, but this is not believed to be important under the conditions investigated. Indications of this include the good prediction of strain rate and pressure trends. In addition, the pressure dependence is like that of other hydrocarbons when they are not strongly influenced by low temperature chemistry. Finally, it was shown for ethane that low temperature chemistry does not qualitatively change the results and only has a small quantitative effect because of the dominance of high temperature pathways during ignition. The qualitative similarity between low temperature mechanisms of different hydrocarbons suggests that this conclusion should also apply to the fuels studied in the present work.

2. Ignition trends of C_1-C_8 hydrocarbons

The similarities noted between the ignition of n-heptane and iso-octane and other hydrocarbons invite a comparison of ignition temperatures under similar conditions. To this end, non-premixed ignition temperatures of C_1-C_8 fuels were measured experimentally, and the results are shown in Fig. 3, revealing that the ignition temperatures have a non-monotonic dependence on the fuel molecule size.

The results in Figure 3 indicate three primary mechanisms that determine the quantitative value of the ignition temperature. The first is the reactivity of the alkyl radical, which depends on the fuel structure. The second is the effect of fuel size on diffusivity. The third is the effect of finite rate kinetics and the complex coupling between reaction and transport for the many intermediate species. The effect of fuel structure is responsible for the high ignition temperatures of methane, iso-butane, and iso-octane. Decreasing fuel diffusivity as fuel size increases is responsible for a rise in ignition temperature from ethane through n-butane. It does not seem likely that this increase will continue indefinitely since this would imply that the large fuel molecules found in many practical fuels would have an extremely high ignition temperature. Indeed, evidence for this increasing trend does not appear beyond n-butane, as the complexities of finite rate kinetics result in a somewhat uniform ignition temperature from n-butane through n-heptane. The variation in ignition temperature for these fuels is less than 20K, which is small relative to the uncertainties associated with determining the actual magnitude of the measured ignition temperatures. This indicates that even though the general finite rate kinetics of hydrocarbon combustion is important, the specific kinetic differences between these fuels, as long as they have similar structures, only have a small effect on ignition temperature.

3. Augmented Reduced Mechanism for methane

A concurrent part of the program has developed a 12-step, sixteen species augmented reduced mechanism (ARM) for methane oxidation. We have previously proposed the concept of developing ARMs for the description of chemical kinetics in complex situations. The premise is that while simplifications of detailed reaction mechanisms must be made, existing reduced mechanisms that typically consist of four to five steps, while simple and physically illuminating, have been shown to have restricted ranges of comprehensiveness in terms of the system dependence on temperature, pressure and concentration. The ARM has shown a high degree of comprehensiveness in its ability to

simulate both the global responses and the temporal and spatial profiles of the temperature and species (both major and minor) of various combustion phenomena over extensive ranges of system parametric variations. In addition, the 12-step mechanism for methane oxidation has been extended to account for NO formation, with a 14-step mechanism to describe the thermal, prompt, and nitrous oxide mechanism, and a 15-step mechanism to include the NH3-related reactions.

4. Characterization of non-reacting turbulent flow

High fidelity experimental data was taken in turbulent non-reacting counterflows. These data are necessary to interpret turbulent *ignition* experiments and to develop and evaluate numerical models for turbulent ignition. It is particularly important to have measurements of counterflows where one jet has a significantly different density than the other jet, as such data is not available in the literature. Counterflowing jets of heated air and cold nitrogen were investigated using a two-component laser Doppler anemometry system to measure velocity moments, spectra, and autocorrelations throughout the flow field. Operating conditions spanned a range of pressures, bulk strain rates, perforated plate configurations, and air temperatures. In addition to measuring velocity moments along the counterflow axis, mean and fluctuating velocity profiles were made in the radial direction at each nozzle exit. These profiles are important for establishing boundary conditions in turbulent flow models. The radial velocity gradients are needed in quasi-one dimensional turbulence models and the profiles themselves are needed for higher dimensional Reynolds stress models and Large Eddy Simulations.

Turbulent power spectra were determined at the burner exits to further characterize the turbulence for a given perforated plate configuration. For example, due to the difficulties associated with generating turbulence in a strongly heated flow, some experiments have been conducted with only one perforated plate, located in the cold flow. The use of this configuration was justified by the observation that the turbulent fluctuations look like those in a turbulent counterflow that has perforated plates in both nozzles. However, a comparison of the power spectra measured at the hot side of the burner, Fig. 4, shows that there is a significant difference between the two cases. The spectrum of the case without a second perforated plate is suggestive of very young, undeveloped turbulence, indicating that it is important to have a perforated plate in both sides of the burner when studying turbulence.

5. Turbulent ignition measurements

Ignition temperatures were measured experimentally for a range of fuel (hydrogen) concentrations, bulk strain rates, and pressures. The effect of hydrogen concentration is shown in Fig. 5. The increasing sensitivity as the fuel concentration decreases and the nearly constant ignition temperature at higher fuel concentrations are qualitatively similar to what was observed in previous non-premixed ignition studies for both hydrogen and other fuels. For all the cases shown, ignition was repeatable and once ignited the turbulent flame persisted, even when the air temperature was subsequently reduced. For lower concentrations, intermittent ignition was observed where above a certain temperature a flame would be repeatedly ignited and extinguished. No hysteresis was observed.

The effect of pressure on ignition is shown in Fig. 6. In light of the laminar hydrogen ignition results, the turbulent data are indicative of a transition between the dominance of second limit chemistry at lower pressures to third limit behavior at the highest pressures. This data will be crucial to evaluating turbulent ignition models since hydrogen has a much higher sensitivity to changes in pressure than other parameters.

6. Modeling of non-reacting turbulent counterflow

In ignition, the amount of heat release *prior to* ignition is small enough that it does not significantly affect the flow or transport properties. As a result, the effects of reaction can be decoupled from the flow such that an equivalent non-reacting turbulent flow field can be solved first. In a subsequent step, the kinetics can be considered using this known flow and the ignition process can be investigated.

Two models were considered to describe the turbulent non-reacting counterflow geometry. A k-ε model was considered initially because of its widespread use together with its simplicity and relatively low computational cost. However, this model was found to greatly overpredict turbulent velocity moments as stagnation is approached. These discrepancies are related to the fact that the k-ε model cannot accurately model the degree of anisotropy present in a counterflow, which distorts the turbulence production term in the governing equations

To address the shortcomings of the k-ε model, a Reynolds stress model was adopted. This model was found to be in reasonable agreement with experimental velocity measurements (Fig. 7) and is a significant enhancement to the k-ε method. This agreement was achieved without adjusting any parameters to fit the calculations to the present experimental data. Information on the scalar mixing fields is not available but the Reynolds stress model showed acceptable agreement with profiles in the literature.
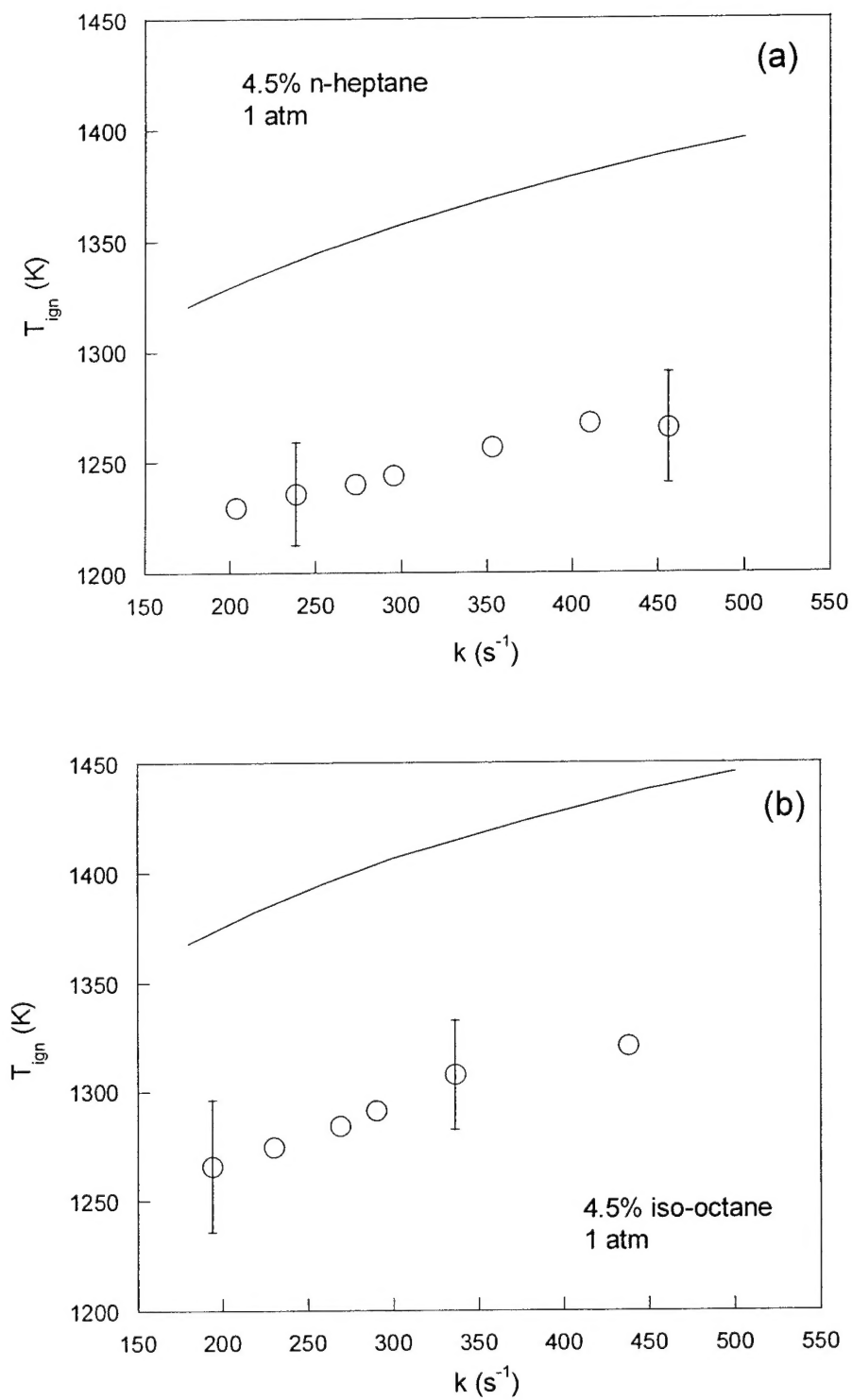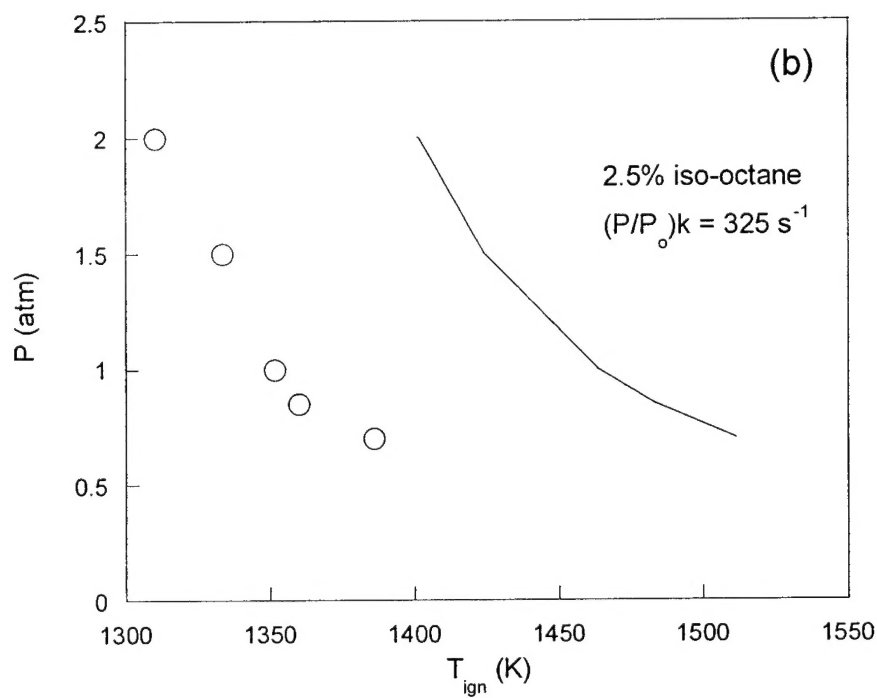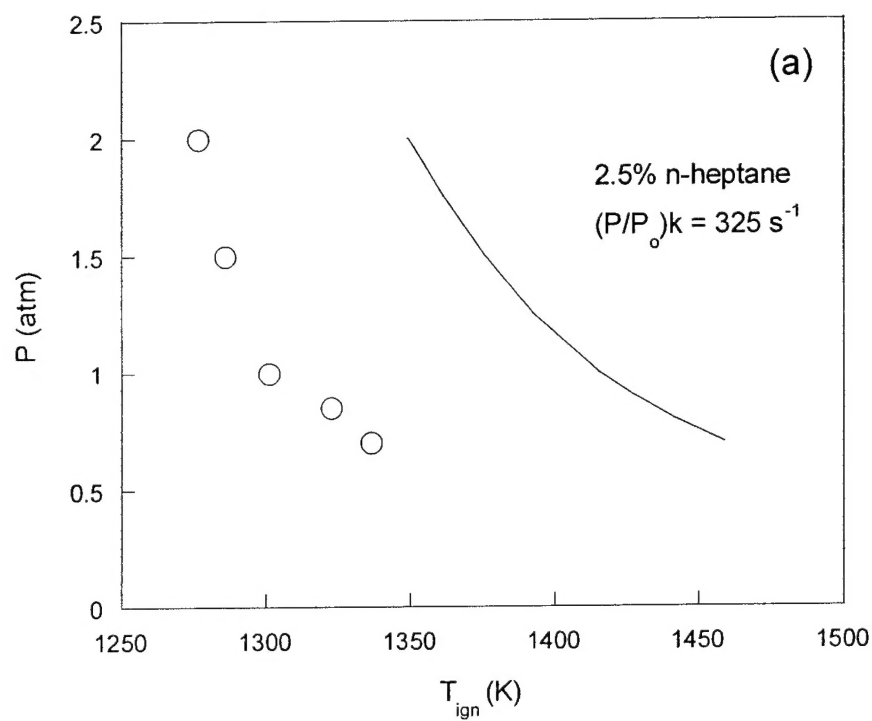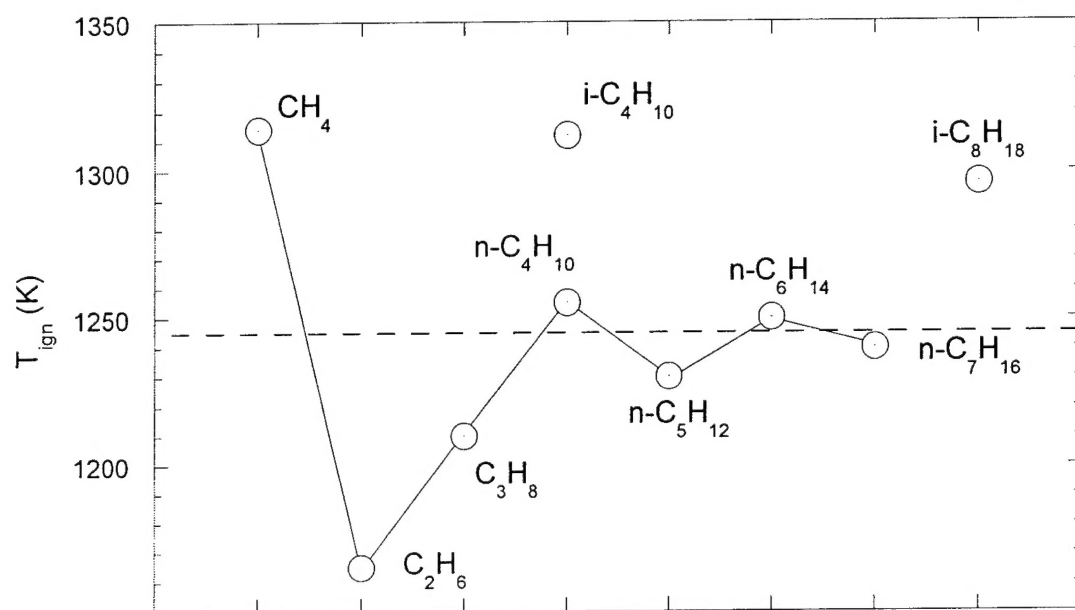
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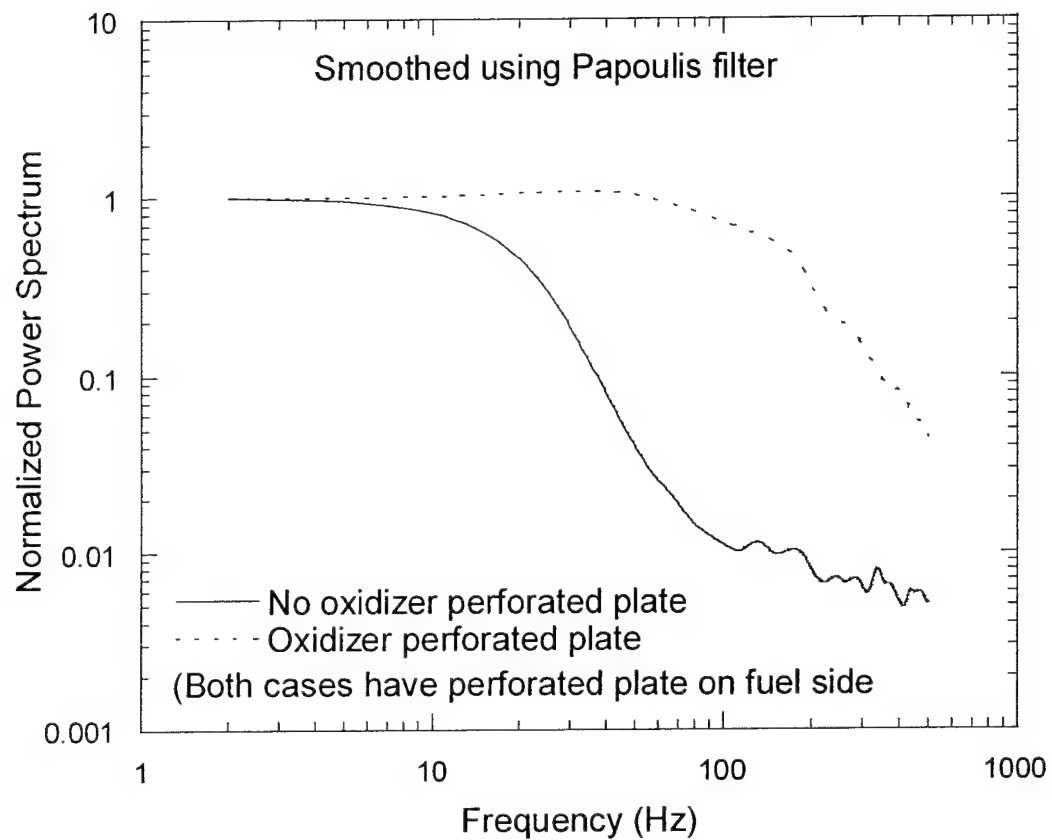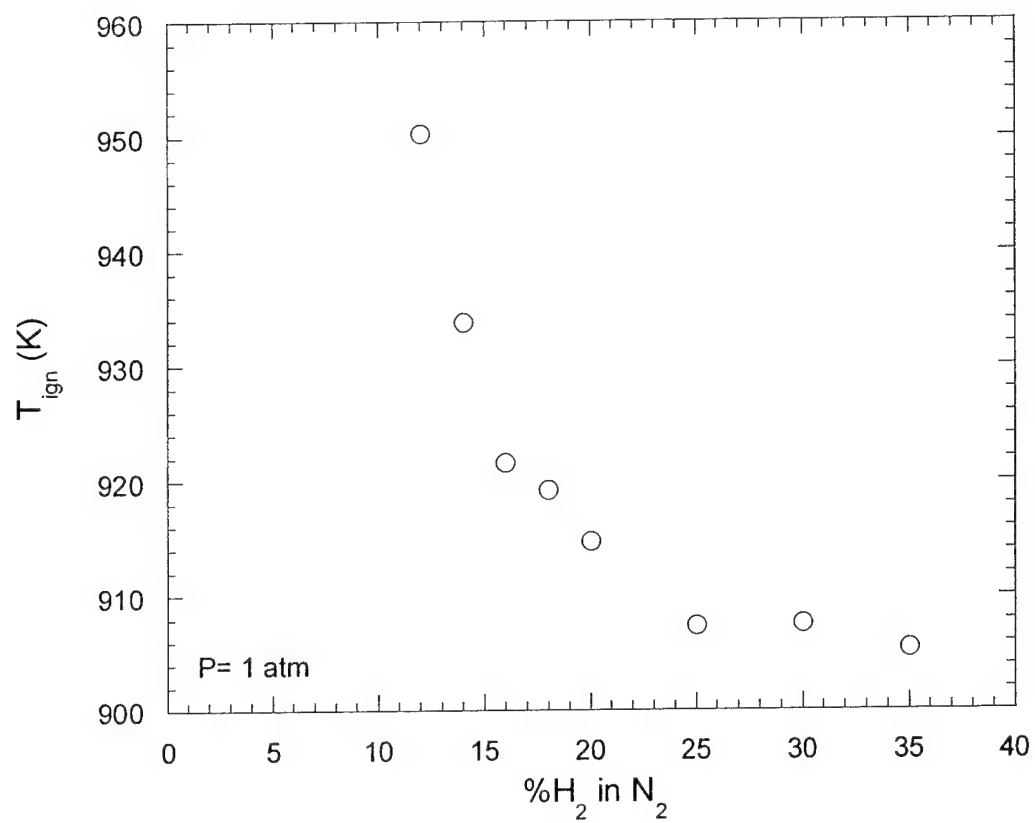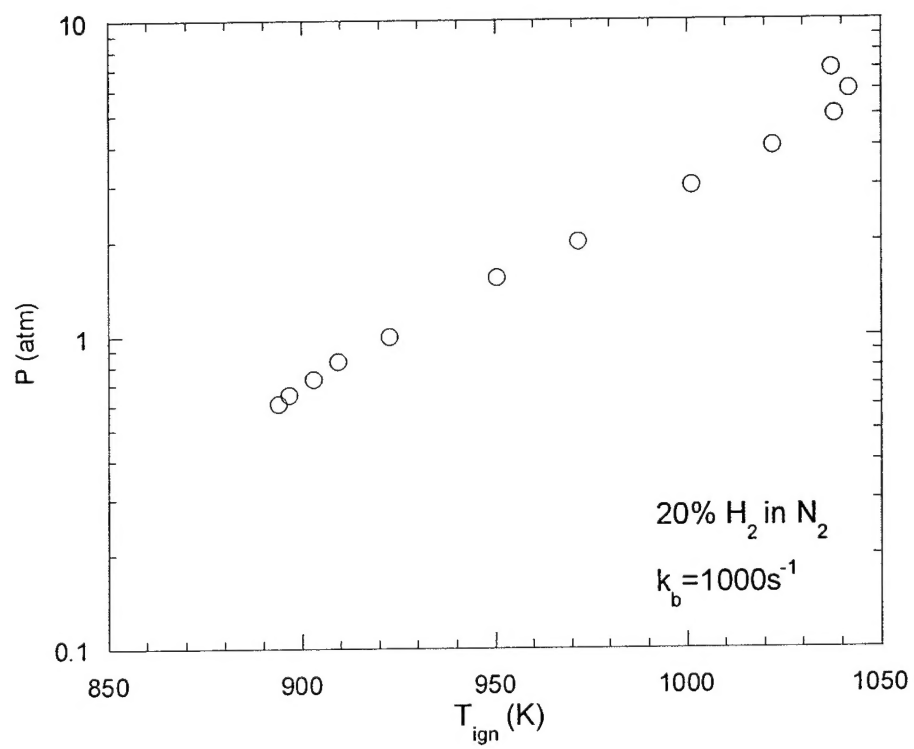Figure 1

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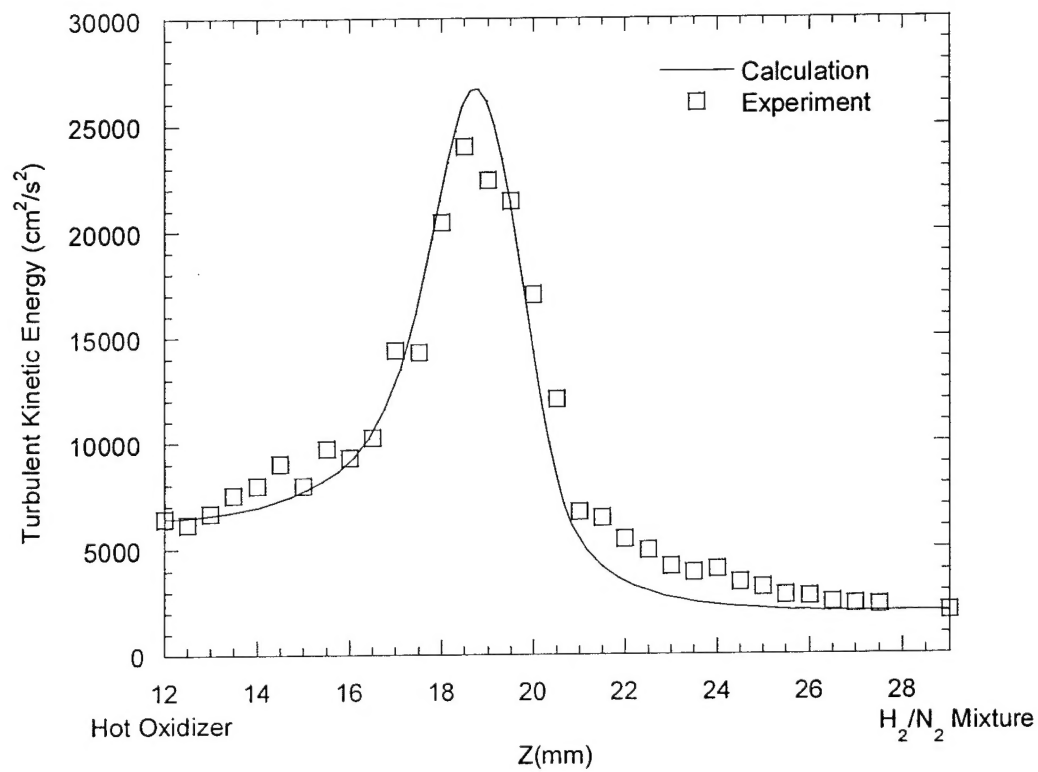Figure 4

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Figure 6

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PUBLICATIONS

JOURNAL AND MAJOR PUBLICATIONS

[1] "Ignition in Nonpremixed Counterflowing Hydrogen versus Heated Air: Computational Study with Skeletal and Reduced Chemistry," by T.G. Kreutz and C.K. Law, *Combust. Flame*, Vol. 114, pp. 436-456 (1998).

[2] "Theory of Radical-Induced Ignition of Counterflowing Hydrogen versus Oxygen at High Temperatures," by B.T. Helenbrook, H.G. Im and C.K. Law, *Combust. Flame*, Vol. 112, pp. 242-252 (1998).

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[6] "Ignition of Ethane, Propane, and Butane in Counterflow Jets of Cold Fuel versus Hot Air Under Variable Pressure," by C.G. Fotache, H. Wang and C.K. Law, *Combust. Flame*, Vol. 117, pp. 777-794 (1999).

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CONFERENCE PROCEEDINGS AND PRESENTATIONS

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[2] "Non-Premixed Ignition of n-Heptane and iso-Octane in a Laminar Counterflow," by J.D. Blouch and C.K. Law, Tech. Meeting of the Eastern States Section of the Combustion Institute, Raleigh, NC, Oct. 1999.

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